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## Direct air capture for concentrating CO<sub>2</sub> for microalgae cultivation

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### Abstract

Microalgae are potentially an attractive renewable resource for feed, food, specialty chemicals and even fuel applications. For sustainability, the required CO<sub>2</sub> for microalgae cultivation must come from a renewable source; preferably ambient air. Often no pure CO<sub>2</sub> is required, but rather a CO<sub>2</sub> enriched air flow with 0.5-2% of CO<sub>2</sub> is applied. In the EU MIRACLES project the objective was to develop on lab scale such a technology, based on regenerative adsorption using solid sorbents. In this work initial results are presented for the developed process, comprising of a radial flow adsorber unit and a moving bed regeneration unit with sorbent circulation between the units. A comparison between batch-wise operation and continuous through-flow of solids in the radial flow reactor showed that the first option was more cost efficient. Sorbent cyclic capacity was below initial estimates based on equilibrium adsorption isotherms. Limiting factors are identified and subject of process improvement efforts. An initial process evaluation, based on energy consumption, is presented.

*Keywords:* direct air capture; sorbent; process; radial flow; enriched air

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### 1. Introduction

Microalgae are, based on their growth rate and versatility, an attractive renewable resource for feed, food, specialty chemicals and possibly even fuel applications. It is known that dosing CO<sub>2</sub> during algae cultivation is required to enhance algae growth rate. For providing CO<sub>2</sub>, locating algae cultivation nearby a large CO<sub>2</sub> emitter, such as stacks from industry or power plants, is a common suggested option. Next to separating and purifying CO<sub>2</sub> from flue gas, often the direct use of the CO<sub>2</sub> rich flue gas is suggested. However, the high temperature of the flue gas and the presence of elevated concentrations of NO<sub>x</sub>, SO<sub>x</sub>, and dust in the flue gas may inhibit algae growth, making direct use of flue gas less attractive. Furthermore, suitable locations and area required for algae cultivation are not necessarily found near large CO<sub>2</sub> emitters. Hence, a separate CO<sub>2</sub> direct air capture (DAC) facility will provide flexibility of location choice for algae cultivation.

For sustainability, the required CO<sub>2</sub> for microalgae cultivation must come from a renewable source, preferably ambient air. In several popular cultivation systems (open ponds, bubble columns, air lift reactors, spray systems etc.) no pure CO<sub>2</sub> is required, but rather a CO<sub>2</sub> enriched air flow with 0.5-2% of CO<sub>2</sub> is used. Within the EU MIRACLES project, the objective was to develop a technology, based on regenerative adsorption using solid sorbents, to concentrate CO<sub>2</sub> from ambient air and demonstrate this at a capacity above 0.5 kg/day. There is a great potential for direct CO<sub>2</sub> capture from ambient air to enhance microalgae cultivation, provided it can technically be realized in an

economical and sustainable manner. It is important to ensure that the  $\text{CO}_2$  captured from the air exceeds the  $\text{CO}_2$  emitted due to energy consumption in the regenerative capture process. Hence, there must be a net  $\text{CO}_2$  uptake.

### 1.1. Process description

In Figure 1, the process concept for regenerative  $\text{CO}_2$  adsorption and the concept for the proposed air sorbent contactor, the so-called radial flow reactor (RFR), as developed in earlier work are shown [2]. In that work a design strategy for a fixed bed type  $\text{CO}_2$  adsorber is developed, based on experimental studies in a fixed bed contactor and using a commercial available sorbent (Lewatit VP OC 1065). This sorbent is also selected for the present study for its good adsorption capacity at ambient conditions and, important for experimental process studies, its availability in larger quantities. As indication, the equilibrium capacity measured in dry air at  $25^\circ\text{C}$  is  $1.1 \text{ mol/kg}$ , which translates to an amine efficiency of 16%. This value is in line with other amine functionalized sorbents [3].

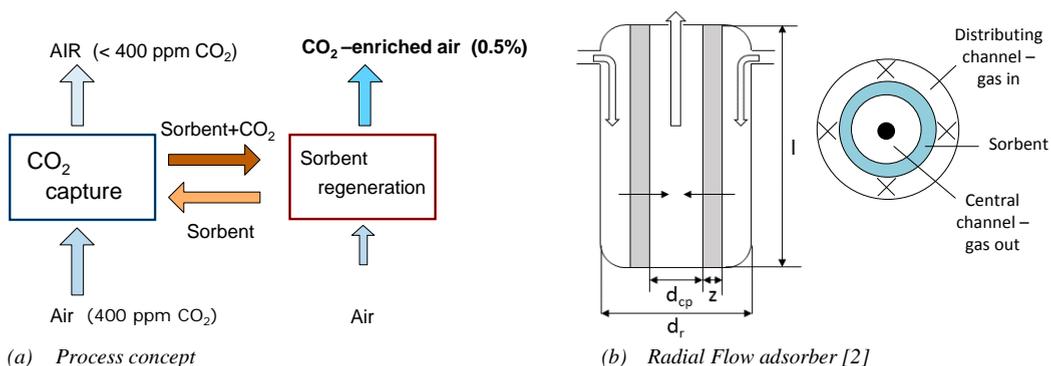


Figure 1 Towards a continuous process for concentrating  $\text{CO}_2$  from ambient air

As embodiment, the use of a radial flow-type of fixed bed reactor was suggested [2] and now built. Alternative gas-solid contactors have also been considered, but were not selected in this project for either reasons of pressure drop or reduced gas-solid contacting (bypassing). The RFR configuration offers a compact design with a small physical footprint, scale-up and scale-out options and, interestingly, the opportunity to implement a moving bed mode of operation, see Figure 2. In this moving bed mode, sorbent is continuously added or withdrawn from the annular space between the two coaxial cylinders. Both the fixed bed mode of operation and the moving bed option will be tested in this work. In the present work the details and performance characterization of the RFR contactor itself and of the pilot unit for direct air capture as a whole are presented.

For regeneration of the sorbent, and the production of the  $\text{CO}_2$  enriched air, different options have been considered. In one of the operating modes the sorbent is regenerated in situ inside the RFR (hence without sorbent recycling), using electrical heating to control temperature. It is however more advantageous, in view of plant capacity and energy efficiency, to do the regeneration step in a separate and to have the sorbent circulating between the two units. Fortunately, the RFR adsorber does allow for (continuous) sorbent inflow and outflow and hence a separate regeneration unit can be implemented. For a separate regeneration unit different options can be thought of, such as a (multistage) fluid bed, moving bed, gas-solid trickle bed, screw conveyer type, again a RFR and so on. Since at the University of Twente already a (multistage) fluid bed and trickle bed contactor have been implemented, it was decided here to test for comparison another type of unit; a slow moving (fluid) bed reactor (MBR). This contactor is sketched schematically in Figure 2. It's main objective is to heat the sorbent to the regeneration temperature and to bring the sorbent (preferably in a counter-current fashion) into contact with the incoming regeneration gas (air) to produce  $\text{CO}_2$  enriched air, to be used in algae cultivation. For demonstration purposes an algae cultivation unit is connected to lab-unit to demonstrate the concept.

## 2. Experimental

The lab scale unit consists of the RFR unit, built as designed [2] and shown in Figure 2 (b), containing 1.7 kg of sorbent. Heart of the  $\pi$ -type RFR are two axial cylinders of a metal woven mesh (porosity 48%), which can be heated electrically by resistance heating. As pressure drop and a uniform flow distribution are important for the performance of the DAC application, these parameters were checked separately. Under the design conditions, the pressure drop for an empty sorbent bed (hence, only the wire meshes) is max. 10 Pa. For the RFR filled with sorbent, the pressure drop varies with gas flow rate from 70-670 Pa when varying the gas flow from around 40 – 320 m<sup>3</sup>/h. Differential pressure measurements over the bed along the axial height showed no measurable differences as function of the position, suggesting a uniform flow distribution.

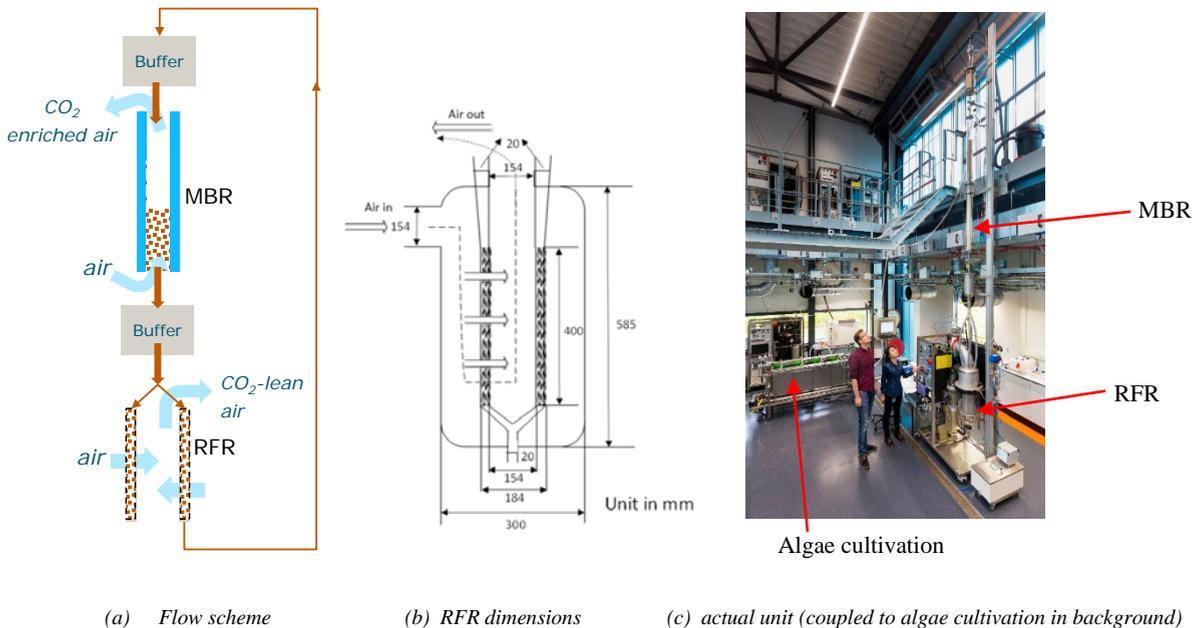


Figure 2 Lab scale unit for producing CO<sub>2</sub> enriched air by direct air capture using solid amine sorbents

The regeneration unit MBR is a double-walled glass tube, heated by hot water, of 60 mm ID and about 2 m long. The MBR and RFR are connected via a sorbent circulation line (20 mm ID) through which the sorbents are transported. This is done using a vacuum facility generating an air flow for sorbent transport, from the bottom outlet of the RFR to a buffer vessel above the MBR, 6 meter higher. There the transported sorbents are separated from the transportation gas by means of a cyclone. The buffer vessels have a capacity of 2.5 kg of sorbent and are thus able to contain the full batch of sorbent coming from the RFR. From the top buffer vessel the sorbents are dosed into the MBR by a calibrated, rotating dosing valve (V1, not shown in Figure 2). Similarly, at the bottom of the MBR a rotating valve is installed to control the outflow of the MBR. At steady state these dosing rates are set equal. In this way a continuous product gas flow can be generated, independent whether or not the sorbent circulation is done batch wise or continuous.

Sorbent circulation was done both continuous and discontinuous. In the discontinuous operating mode, a ball valve at the bottom of the RFR is opened completely to allow the sorbent to flow out of the adsorber into the riser section to transport the (now: CO<sub>2</sub> loaded-) sorbent to the cyclone at the top of the set-up. The vacuum facility used generating the riser gas was able to transport the 1.7 kg of sorbent within 2 minutes to the top buffer vessel. As ambient air is used for sorbent transportation, possible CO<sub>2</sub> pre-desorption in the riser can be neglected. In the

continuous sorbent circulation mode rotating valve V1 is moved from its position above the MBR to the outlet of the RFR at the bottom end of the setup.

The air flow through the RFR and MBR are controlled by a fan (0-300 m<sup>3</sup>/h) and a mass flow controller (0-100 NL per minute) respectively. CO<sub>2</sub> concentrations and temperatures are measured. Additionally, humidity meters are installed in the ingoing and outgoing air flows to assess the extent of water co-adsorption.

For operation of the entire unit the following process options were tested:

- (1) batch-wise: in-situ adsorption and desorption, where the RFR acts as a shallow fixed bed;
- (2) continuous adsorption in the RFR (the sorbent is slowly moving down inside the RFR), with continuous sorbent regeneration in the MBR
- (3) semi-batch wise adsorption in the RFR, with continuous desorption in the MBR

For demonstration purposes only, during two weeks an illuminated photobioreactor for algae cultivation was installed next to the DAC facility to demonstrate the cultivation on a DAC-generated CO<sub>2</sub>-enriched air stream. The species cultivated was *Nannochloropsis sp.*

### 3. Results

#### 3.1 Process conditions

Since optimizing the conditions of the regenerator is limited by the conditions acceptable in view of sorbent degradation [4], additional sorbent stability tests (cyclic vs. continuous exposure, effect of temperature and long term exposure tests (up to 1000 h) were carried out (results not shown here). It was found that oxidative degradation of the sorbent above 70°C led to unacceptable degradation rates. Therefore the operating temperature for the MBR is kept below this value.

Based on the equilibrium adsorption isotherms for CO<sub>2</sub> in dry gas, the cyclic capacity of the sorbent could be up to 1 mole/kg, based on adsorption (from air) at 293 K and desorption (in air) at 333 K. The possible effect of water co-adsorption on the CO<sub>2</sub> capacity [5] is neglected in this estimation. This potential cyclic working capacity was considered to be sufficient. A typical sorbent circulation velocity through the calibrated rotary valves into- and out of the regenerator is 0.65 g/s. In combination with the maximum (equilibrium based) cyclic capacity a total production capacity of the unit of around 2.4 kg CO<sub>2</sub> per day can be calculated. With this, the potential maximum capacity is well above the targeted production capacity of 1 kg/day.

#### 3.2 Batchwise operation of RFR as adsorber and desorber

For producing CO<sub>2</sub> enriched air, first the RFR adsorber was used for both the adsorption and desorption step, see process option (1) in the Experimental section. A typical result is presented in Figure 3, in which an adsorption time of 43 minutes and a regeneration time of 80 minutes were implemented. In this figure, first the sorbent regeneration phase (using 4.5 m<sup>3</sup>/h of air as purge gas) and subsequently the sorbent re-adsorption phase are presented. It can be seen that the sorbent bed is heated relatively fast (5 minutes) to 50°C after which it much slower heats to finally 65°C. The Figure further shows, the instantaneous CO<sub>2</sub> outlet concentration as well as the time-averaged mixing cup concentration.

In the experiment presented in Figure 3, the cyclic sorbent capacity realized was 0.75 mole/kg, as determined by the amount adsorbed in the following adsorption step. For comparison, a second run is done using regeneration in hot (80°C), CO<sub>2</sub>-free, in a nitrogen flow of 3 m<sup>3</sup>/h for 16 hours. This prolonged regeneration test in nitrogen yielded a slightly higher cyclic capacity of 0.80 mole/kg. This implies that the regeneration in air, as shown in Figure 3,

yielded nearly the same cyclic capacity (94%), as when using  $N_2$  under the prevailing conditions. The presence of  $CO_2$  or water in the incoming regeneration air is thus not significantly disturbing the regeneration process.

The difference of the obtained 0.75 mole/kg (or 0.8 mole/kg for  $N_2$ ) with the earlier mentioned maximum cyclic capacity of 1.0 mole/kg can be due to one or more different effects. First of all, the applied adsorption time ( $t_{ads}$ ) of 43 minutes is approximately equal to the stoichiometric time ( $t_{sto}$ ). This stoichiometric time is the time minimally needed to saturate the sorbent bed when all incoming  $CO_2$  would be adsorbed. This parameter was also used as scale-up parameter in the design strategy for the unit. Earlier work using fixed bed experiments has shown that when  $t_{ads} = t_{sto}$ , the degree of sorbent saturation ( $\eta_s$ ) is around 57%, for a similar gas velocity and bed thickness as used in the RFR [2]. In other words, the adsorption phase is not long enough to reach full sorbent saturation. It can be calculated, when regeneration in hot nitrogen for 16 h is assumed to be complete, that the equilibrium adsorption capacity then would amount  $0.8/0.57 = 1.4$  mole  $CO_2$ /kg at the prevailing adsorption temperature (20–22°C) and relative humidity (55–60%). This estimated equilibrium capacity for  $CO_2$  in humid air is well in line with the limited data set available in literature, from which an adsorption capacity of around 1.55 ( $\pm 0.1$ ) mole/kg is estimated, based on Figure 8 in the work by Veneman et al. [5].

When assuming 57% adsorption efficiency and an equilibrium adsorption capacity for  $CO_2$  at the adsorption temperature and relative humidity during the test of ca. 1.55 mole/kg, then the maximum  $CO_2$  loading of the sorbents in the RFR contactor is around 0.88 mole/kg. With the above estimated 94% regeneration efficiency, the cyclic capacity to be expected is then 0.83 mole/kg (to be compared with the experimental determined 0.75 mol/kg value). Considering the uncertainties in the estimated values above and in the measured parameters (as relative humidity, flow rates and gas phase concentrations), one might argue that the remaining deviation is within the level of accuracy. Alternatively, this remaining difference of 0.08 mole/kg with the experimental obtained 0.75 mole/kg might be caused by a fraction of the sorbent content of the RFR not being active (less than 10%), or an decreased efficiency of the regeneration step due to e.g. heat transfer limitations. It should further be realized that when using humid air (4.5  $m^3/h$ ) instead of dry nitrogen (3  $m^3/h$ ) for same regeneration experiment the actual heat demand is larger and the actual bed temperature may be slightly different. Considering the poor conductivity of the sorbent particles, it might be that the heat transfer rate was partly limiting the degree of regeneration. The consistency of the experimental data for the cyclic capacity in comparison with the expected ones (0.75 vs. 0.83 mol/kg), is however considered sufficient and the remaining differences were not further analysed.

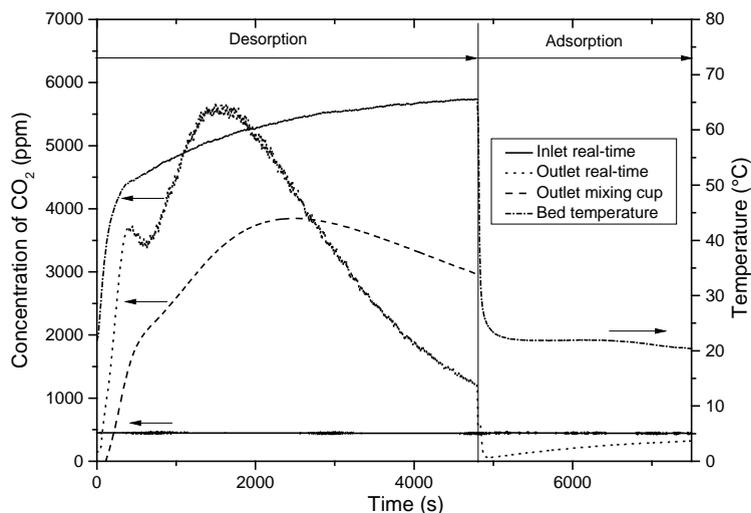


Figure 3.  $CO_2$  concentration profiles for RFR outlet during the desorption phase in air (4.5  $m^3/h$ ) and subsequent adsorption phase.

### 3.3 RFR as continuous adsorber

To produce a CO<sub>2</sub> enriched product stream with constant concentration a system with continuous desorption in a separate (MBR) regenerator is envisaged. It was therefore tested whether the RFR adsorber, operated batch wise as described above in section 3.2, could also be used as continuous adsorber. For this a continuous sorbent flow was entering the RFR at the top. From loading/unloading tests of the RFR it is known that the solids are free flowing and distribute nicely over the entire circumference of the annular space between the coaxial cylindrical wire meshes. The sorbent flow rate through the RFR is set by a rotating valve, located at the bottom exit of the RFR.

For a direct comparison with the batch-wise operation of the RFR, the sorbent flow rate was set at 0.6 g/s to achieve the same residence time of the sorbents inside the RFR as the duration of the adsorption phase in the corresponding batch experiment (Figure 3). To make data-interpretation for this comparison straight forward, first the regenerator and buffer vessels were filled with completely unloaded sorbent (because regenerated in hot N<sub>2</sub> for several hours) and only the contacting efficiency of the RFR as continuous adsorption device is tested.

In the achieved steady state situation for the RFR as adsorber, the outgoing air flow of the RFR contained still 65% of its original (incoming) CO<sub>2</sub> content; hence the CO<sub>2</sub> removal fraction ( $\eta_G$ ) is 35%. At  $t_{ads}=t_{sto}$ , the sorbent saturation efficiency ( $\eta_S$ ) and gas removal efficiency ( $\eta_G$ ) are by definition equal [2]. Hence, for the batch-wise operation in Figure 3,  $\eta_G$  equals  $\eta_S$  and amounts 57%. It is now clear that the removal efficiency  $\eta_G$  for the batch-wise operation (57%) is much higher than for the continuous sorbent flow mode of operation (35%).

A plausible explanation for this reduced removal efficiency when operating in the continuous mode is the decrease of the (now: cross-flow) contacting efficiency between incoming air and the sorbents. Apparently, the average particle-particle distance in the moving bed inside the RFR is slightly larger, leading to some bypassing of the gas phase. An alternative explanation (or additional contribution) could be that in the lower section of the RFR the fresh incoming air is in contact with partly loaded sorbent. If this affects the adsorption kinetics strongly, then the air in this section will be less depleted. Depending on the dependency of the adsorption kinetics on the particle loading, this might also contribute to the observed difference.

Rather than analysing this difference in detail, an alternative mode of operation was designed. It was found that the entire sorbent content of the RFR can be transported rather quickly (within 1-2 minutes) from the adsorber to the top buffer vessel. In comparison with the used adsorption times in the RFR (typical 45-60 minutes), the time for sorbent transport is marginal. By placing the sorbent-dosing valves above and below the regeneration unit, the RFR can still be used in a batch-wise mode, with fast sorbent transport to the top buffer vessel, while the regenerator operates in a continuous fashion, yielding a constant product gas of CO<sub>2</sub> enriched air (option (3), see next section).

### 3.4 Semi-batch wise adsorption in RFR and continuous regeneration in MBR

For process option (3), the RFR is operated batch wise, but (as enabled by the intermediate storage buffers) the regeneration in the MBR is continuous. Figure 4 shows the outlet concentration in the product gas for the Proof of Concept run. In this Figure the start-up phase (first hour) and the subsequent continuous operation of the unit over several hours can be recognized. The conditions in the regenerator (temperature of 60°C and air purge flow rate of 1.2 m<sup>3</sup>/h) are set in such a way that a product gas with (on average) a concentration of 1% of CO<sub>2</sub> is obtained. From the data, the calculated production capacity of the facility is around 0.57 kg CO<sub>2</sub>/day. With this, the targeted production capacity and CO<sub>2</sub> product gas concentration were both achieved!

The results show, however, also that for the presented experimental run the corresponding cyclic capacity of the sorbent is around 0.23 mol/kg, which is significant lower than the above discussed cyclic capacity of 0.75 mol/kg, which was achieved when using the unit in process option (1). In that latter option, with the RFR as fixed bed for both the adsorption and desorption step, the cycle time of the RFR is equal to the sum of  $t_{ads}$  and  $t_{des}$  (for Figure 3

resp. 43 and 80 minutes), which leads overall to a fairly similar capacity of the unit of 0.66 kg CO<sub>2</sub> per day. Hence, the higher cyclic capacity for process option (1) does not lead to proportional higher system productivity, as the time on stream for the RFR in the adsorption mode is much lower. A clear advantage of process option (3) is the more constant product quality; compare hereto the CO<sub>2</sub> concentration fluctuations in Figures 4 and 3.

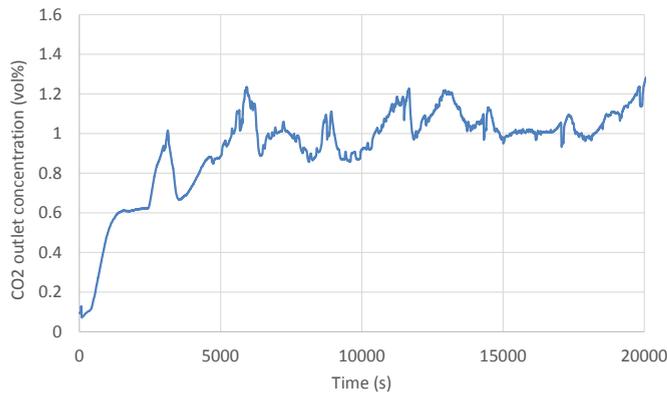


Figure 4. MBR outlet concentration of CO<sub>2</sub> in the CO<sub>2</sub> enriched air product gas.  
Conditions:  $T(MBR) = 333\text{ K}$ ;  $\Phi_{v, MBR} = 1.2\text{ m}^3/\text{h}$ ;  $\Phi_{s, MBR} = 0.65\text{ g/s}$

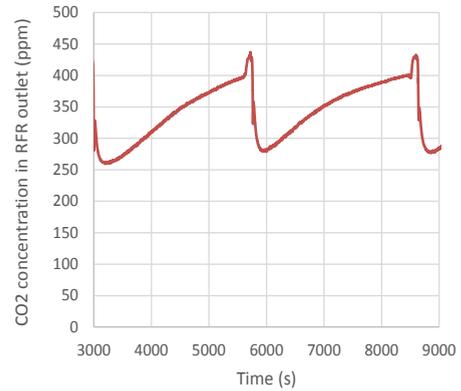


Figure 5 RFR outlet concentration for CO<sub>2</sub>  
Conditions:  $T=295\text{ K}$ ,  $\Phi_G = 188\text{ m}^3/\text{h}$ ,

The reduced cyclic sorbent capacity, in comparison with option (1), can not be due to the adsorption step, as the conditions and duration of the adsorption phase are identical. It is therefore clear that the overall lower cyclic adsorption capacity is due to the incomplete regeneration in the MBR. This is also evidenced by the CO<sub>2</sub> concentration profile of the outgoing air in the RFR, which does not show the low outlet concentrations as in Figure 3 (at approximately  $t = 5000\text{ s}$ ), but does not go below 250 ppm, see Figure 5. Note that the peaks in Figure 5 are related to the emptying/filling of the sorbent bed in the RFR and exceed 400 ppm significantly as the CO<sub>2</sub> concentration in the lab at the time of measurement was around 436 ppm.

### 3.5 DAC for algae cultivation

Option (3) was select for the production of a continuous stream of CO<sub>2</sub> enriched air for the cultivation of the marine microalgae species *Nannochloropsis sp.* in a sparged photobioreactor, illuminated with LED's. For illustration only, a rough indicative comparison of the initial growth rates, with and without the application of the produced CO<sub>2</sub> enriched air from the novel DAC unit is presented in Figure 6.

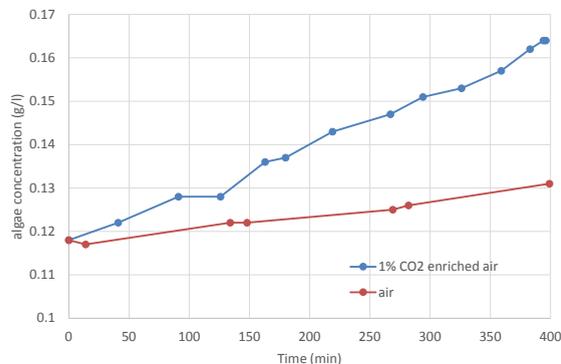


Figure 6. Algae growth rate using air (red line) and CO<sub>2</sub> enriched air (blue line)

## 4. Discussion

The three different process options lead to different production capacities and different cyclic capacities for the sorbent used. From a plant capacity point of view, the options (1) and (3) are not significantly different. Option (2) is not (yet) evaluated in detail. The maximum capacity for option (2), as determined by adsorption using complete regenerated sorbent (see section 3.3), is almost double that of options (1) and (3), namely 1.25 kg/day. But since no run with integrated regeneration in the MBR was conducted, this number can not directly be compared with the result for option (3). A capacity of 1 kg CO<sub>2</sub> per day in this unit translates to a carbon fixation rate of 10,000 ton C/ha yr, which exceeds the areal C fixation rate of biomass by a few orders of magnitude.

Moreover, the results in Figure 4 show that the CO<sub>2</sub> concentration in the product gas is around 1.0 % in air. According to the adsorption isotherm (for dry gas), the equivalent CO<sub>2</sub> equilibrium capacity on the sorbent is approximately 1.0 mole/kg. The loaded sorbent entering the MBR should therefore have a sorbent loading exceeding this value by (at least) the measured cyclic capacity. Hence, for option (3) and the results presented in section 3.4 this means the loaded sorbent should have a loading of at least  $q_{ads} = 1.23$  mol/kg. As the analysis in section 3.2 pointed out, such a sorbent loading is not feasible under the test conditions, when starting with a completely lean sorbent ( $q = 0$  mol/kg). This underlines the occurrence of incomplete regeneration in the regenerator which one might want to improve upon. However, it also shows that complete sorbent regeneration is probably not even desired (depending on the required CO<sub>2</sub> concentration in the product gas).

A simple analysis for a series of consecutive adsorption/desorption cycles, taking into account that around 50% of the driving force for adsorption is reduced in the adsorption phase ( $\eta_{ads} = 57\%$  was found in section 3.2) and assuming a constant regeneration fraction ( $\eta_{des}$ ), shows that steady state is achieved after 3-4 cycles, in line with Figure 4. From comparison with the data in section 3.4 it was estimated that this regeneration efficiency ( $\eta_{des}$ ), defined as the reduction of the sorbent loading in excess of the loading at equilibrium with the incoming air purge, as evaluated at regeneration temperature, is only around 20%. Figure 6 shows the values for the cyclic capacity ( $\Delta q$ ) and outlet concentration of CO<sub>2</sub> according to this method. Complicating factors such as the lower temperature of the incoming purge gas have been neglected. This relatively low regeneration efficiency factor (to be compared with e.g. the 94% as determined in section 3.2) is due to the MBR performance, most likely with respect to heat transfer. Further study to improve heat transfer (heated wall to bed of sorbents) and an increased sorbent residence time in the MBR could reduce this limitation. If the regeneration efficiency can be improved to e.g. 50%, then (according to the same estimation method as used for Figure 6) the cyclic sorbent capacity increases to around 0.5 mol/kg, at the expense of a small reduction in the outlet concentration to around 0.9 vol.%

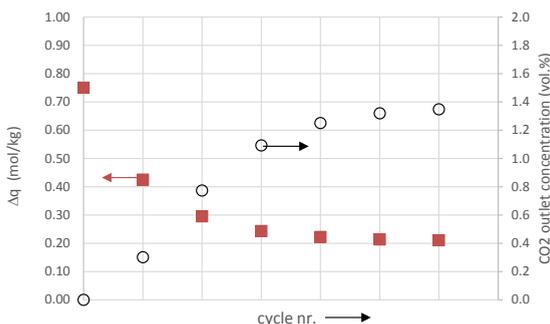


Figure 6. Estimated cyclic capacity and CO<sub>2</sub> outlet concentration Assuming  $\eta_{ads} = 50\%$  and  $\eta_{des} = 20\%$

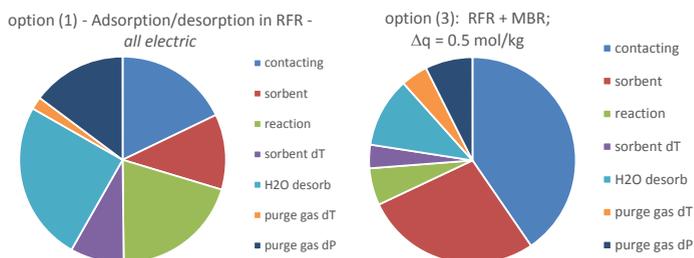


Figure 7. Preliminary operating cost distribution for option (3) assumptions: 0.08 €/kWh electr.; 3 €/GJ thermal;  $q_{H_2O} = 5$  mol/kg; 70% purge gas heat recovery, 5 mbar pressure drop (air)

**DISCLAIMER:** These premature costs figures in Figure 7 and text below contain a significant level of uncertainty (e.g. in energy consumption, water co-adsorption effects, heat integration possibilities, sorbent life etc.) and the prices for heat and electricity may vary significantly in place and time. Furthermore, capital costs are not included, which may significantly alter the overall cost comparison between the different options.

A preliminary cost evaluation for the process options (1) and (3) showed that the operating costs depend heavily on the cyclic capacity, especially in combination with water co-adsorption and on purge gas heat integration. For option (1) and the conditions reported in section 3.2, the total operating costs are estimated at the level of 160 €/ton CO<sub>2</sub> (for indicated prices, see caption Figure 7). For option (3) the costs are significant lower (around 80 €/ton CO<sub>2</sub>) when the MBR can be improved to realise the above mentioned 0.5 mol/kg as cyclic capacity. The cost distribution presented in Figure 7 is based upon this latter assumption. With increasing cyclic capacity the operating costs are even further reduced. For the cyclic capacity of 0.23 mol/kg, as observed in the presented experiments in Figures 4 and 5, this value is around 120 €/ton. Figure 7 indicates, above all, that there is not a single dominant cost factor.

For the net CO<sub>2</sub> footprint, there is a strong relation with the energy used to drive the process. If the energy is renewable (e.g. electricity from solar PV with an estimated life cycle average CO<sub>2</sub> footprint of 50 g/kWh<sub>el</sub> and 7.5 kg CO<sub>2</sub> per GJ solar thermal heat), the net CO<sub>2</sub> uptake is around 0.9 kg CO<sub>2</sub> per kg CO<sub>2</sub> captured.

## 5. Conclusions

In this work preliminary test results for a novel direct air capture unit, based on regenerative CO<sub>2</sub> adsorption with a supported amine sorbent, are presented. The unit, consisting of a radial flow reactor for adsorption/desorption and a moving bed regenerator, was used in three different modes of operation. These include full batch operation in the RFR to continuous sorbent circulation between the RFR adsorber and MBR. The tested configurations were able to meet the targeted production capacity of 0.5 kg CO<sub>2</sub>/day and producing a product gas with 0.3-1% CO<sub>2</sub>. From the cyclic sorbent capacities determined for the different options, it was found that at the temperature tested, the regeneration process in the MBR is limiting the unit performance with respect to its capacity. However, this incomplete regeneration simultaneously leads to a higher CO<sub>2</sub> concentration in the product gas.

Among the options, the semi-batch operation of the RFR with continuous regeneration in the MBR was selected as most promising for the relative high good air-sorbent contacting and producing a continuous flow of CO<sub>2</sub> enriched air at constant quality. A preliminary cost evaluation showed that there is no single dominating factor; factors as pressure drop (gas-sorbent contacting), sorbent costs, cyclic capacity and water co-adsorption all play a role. Further tests, modeling and optimization is needed to optimize the process, especially the regeneration step. The next step is then designing and testing at an even larger capacity to get a better view on scalability and on capital costs involved.

### Nomenclature

T	temperature	(K)
t	time	(s)
t <sub>sto</sub>	stoichiometric contact time	(s)
q	sorbent CO <sub>2</sub> loading	(mol/kg)
Φ	flow rate	(m <sup>3</sup> /h) or (kg/h)
η <sub>G</sub>	gas removal efficiency	(-)
η <sub>s</sub>	sorbent saturation efficiency	(-)

### Sub- and superscripts

ads	adsorption (-conditions)
des	desorption (-conditions)

## References

- [1] Brilman W, Garcia Alba L, Veneman R, Capturing atmospheric CO<sub>2</sub> using supported amine sorbents for microalgae cultivation, 2013, *Biomass Bioenergy*, 53, 39-47
- [2] Yu Q., Brilman D.W.F., Design strategy for CO<sub>2</sub> adsorption from ambient air using a supported amine based sorbent in a fixed bed reactor, 2017, *Energy Procedia*, 114, 6102-6114
- [3] Chaikittisilp W, Khunsupat R, Chen T.T., Jones C.W., Poly(allylamine)-Mesoporous Silica Composite Materials for CO<sub>2</sub> Capture from Simulated Flue Gas or Ambient Air. *Industrial and Engineering Chemistry Research*, 2011, 50, (24), 14203-14210
- [4] Yu Q., de la Puente Delgado J., Veneman R., Brilman DWF, Stability of a Benzyl Amine Based CO<sub>2</sub> Capture Adsorbent in View of Regeneration Strategies, 2017, *Ind.Eng.Chem.Res.*, 56, 3259-3269
- [5] Veneman R., Frigka N., Zhao W., Li Z., Kersten S., Brilman W., Adsorption of H<sub>2</sub>O and CO<sub>2</sub> on supported amine sorbents, 2015, *Int. J.Greenhouse Gas Control*, 41, 268-275